

Competition of local-moment ferromagnetism and superconductivity in Co-substituted EuFe_2As_2

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Abstract. In contrast to SrFe_2As_2 , where only the iron possesses a magnetic moment, in EuFe_2As_2 an additional large, local magnetic moment is carried by Eu^{2+} . Like SrFe_2As_2 , EuFe_2As_2 exhibits a spin-density wave transition at high temperatures, but in addition, the magnetic moments of the Eu^{2+} order at around 20 K. The interplay of pressure-induced superconductivity and the Eu^{2+} order leads to a behavior which is reminiscent of re-entrant superconductivity as it was observed, for example, in the ternary Chevrel phases or in the rare-earth nickel borocarbides. Here, we study the delicate interplay of the ordering of the Eu^{2+} moments and superconductivity in $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$, where application of external pressure makes it possible to sensitively tune the ratio of the magnetic (T_C) and the superconducting ($T_{c,\text{onset}}$) critical temperatures. We find that superconductivity disappears once $T_C > T_{c,\text{onset}}$.

1. Introduction

The discovery of high-temperature superconductivity in the iron-based superconductors has stimulated an enormous interest in the study of this new class of materials. One peculiar finding is the interplay of the local $4f$ moments of the Eu^{2+} ions and superconductivity in EuFe_2As_2 under pressure, which is reminiscent of re-entrant superconductivity [1, 2]. Like the ($A=\text{Ca}, \text{Sr}, \text{Ba}$) members of the $A\text{Fe}_2\text{As}_2$ family, EuFe_2As_2 exhibits a spin-density wave (SDW) transition around $T_0 = 190$ K related to the Fe_2As_2 layers, but in addition the magnetic moments of the localized Eu^{2+} order at $T_N = 19$ K [3, 4]. EuFe_2As_2 has a similar crystallographic and electronic structure compared to that of SrFe_2As_2 [3]. Therefore, SrFe_2As_2 can be considered a non- f homolog of EuFe_2As_2 . In $\text{SrFe}_{2-x}\text{Co}_x\text{As}_2$ Co-substitution in the Fe_2As_2 layer stabilizes a superconducting (SC) phase ($0.2 \lesssim x \lesssim 0.4$) leading to the expectation that Co-substitution in EuFe_2As_2 does the same, which in the meantime has been confirmed experimentally [6, 7].

In this paper we present a pressure study on $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$ by means of electrical-resistivity (ρ) and ac-susceptibility (χ_{ac}) measurements on polycrystalline samples. In $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$ in our accessible pressure range we can sensitively tune the magnetic ordering temperature T_C from $T_C < T_{c,\text{onset}}$ to $T_C > T_{c,\text{onset}}$ allowing us to study the peculiar interplay of $4f$ magnetism and superconductivity.

2. Experimental Details

The polycrystalline samples of $\text{EuFe}_{2-x}\text{Co}_x\text{As}_2$ were synthesized by sintering stoichiometric amounts of the precursors EuAs , Fe_2As and Co_2As . The use of precursors minimizes the

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elemental impurities of As and Fe. X-ray diffraction measurements confirmed the BaAl_4 type structure (space group $I4/mmm$) for all the samples. Electrical resistance and ac susceptibility were measured using an LR700 resistance/mutual inductance bridge (Linear Research). A miniature compensated coil system placed inside the pressure cell was utilized for the χ_{ac} experiments. Temperatures down to 1.8 K were reached using a physical property measurement system (PPMS, Quantum Design). Pressures up to 3 GPa have been achieved in a double-layer piston-cylinder type pressure cell with silicone fluid as pressure transmitting medium. The SC transition of Pb, which served as a pressure gauge, remained sharp at all pressures indicating good hydrostatic conditions.

3. Experimental Results

Figure 1 shows the resistance normalized to the value at room temperature, $R/R_{300\text{K}}(T)$, for $\text{EuFe}_{2-x}\text{Co}_x\text{As}_2$ ($x = 0, 0.1, 0.2$, and 0.3). In EuFe_2As_2 $R/R_{300\text{K}}(T)$ exhibits two distinct features at $T_0 = 195$ K and $T_N = 12.5$ K corresponding to the SDW instability and the ordering of the localized Eu^{2+} moments, respectively, in good agreement with literature [3, 4]. Upon Co substitution T_0 decreases rapidly to 103 K for $x = 0.1$. At $x = 0.2$ no signature of the SDW transition is visible in the $\rho(T)$ data anymore. $T_{N,C}$ exhibits a weak concentration dependence. In the concentration range between $x \approx 0.1 \div 0.2$ the type of magnetic order possibly changes from antiferromagnetic (AFM) to ferromagnetic (FM). In $\text{EuFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ a sharp drop of the resistance below ≈ 10 K indicates the onset of superconductivity. This observation is consistent with studies on single crystals [6]. However, there a higher T_c and reentrant-like SC behavior was reported. For $x \leq 0.1$ and $x \geq 0.3$ no signature of superconductivity is visible in our data evidencing a more narrow SC region than in the non- f homolog system $\text{SrFe}_{2-x}\text{Co}_x\text{As}_2$ [5].

In the following, we will focus on the results of our pressure study on $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$. $\rho(T)$ for different pressures is shown in Fig. 2. Upon applying pressure the anomaly indicating the SDW transition at T_0 broadens and shifts rapidly to lower temperatures. At 1.61 GPa no feature related to T_0 can be resolved in $\rho(T)$ anymore suggesting that the SDW order is already suppressed at this pressure. At low temperatures, displayed in Fig. 3a, a kink in $\rho(T)$ at 14.8 K and 16.9 K indicates the ordering of the Eu^{2+} moments at 0 and 0.43 GPa, respectively [9]. At 1.01 GPa a first signature of a SC transition becomes evident: $\rho(T)$ drops sharply below $T_{c,\text{onset}} = 24.2$ K followed by a small maximum indicating the magnetic ordering of the Eu^{2+} and a further decrease in $\rho(T)$ on lowering the temperature. This behavior is similar to that

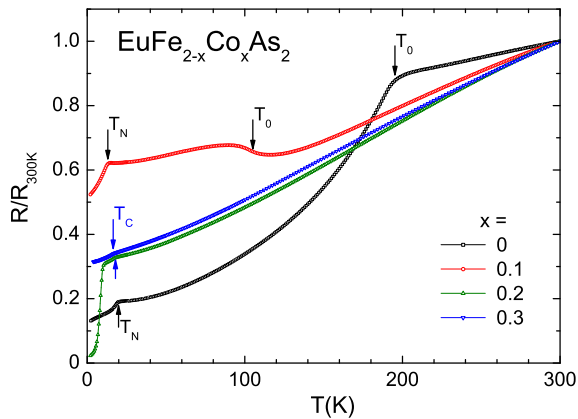


Figure 1. Electrical resistivity normalized to the value at room temperature ($R/R_{300\text{K}}$) for $\text{EuFe}_{2-x}\text{Co}_x\text{As}_2$.

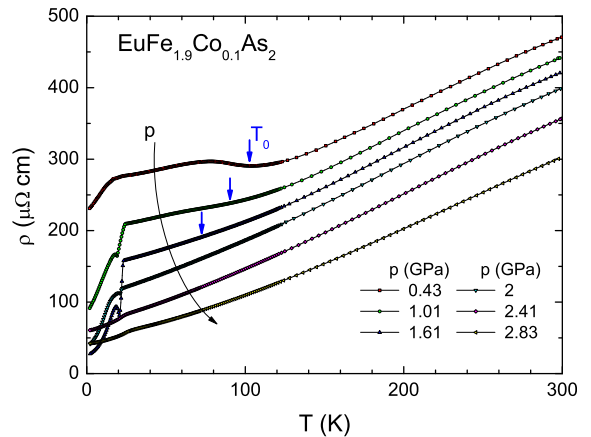


Figure 2. $\rho(T)$ of $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$ at different external pressures.

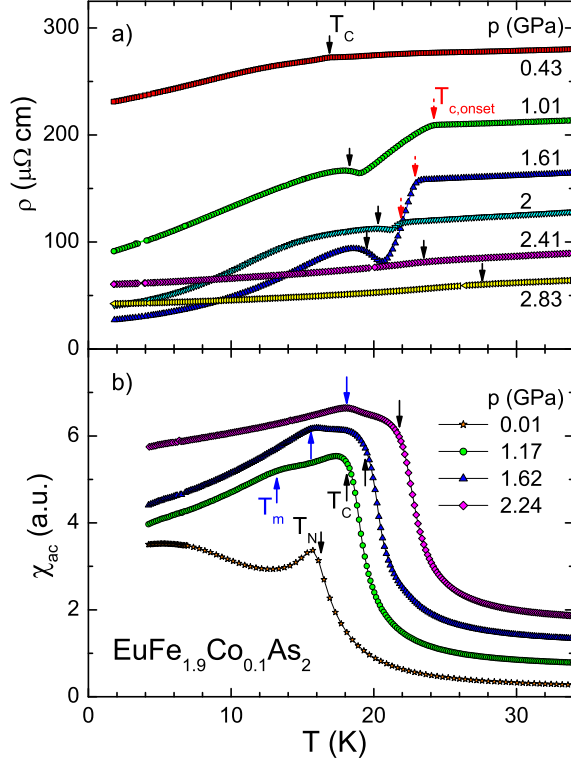


Figure 3. (a) Low-temperature resistivity and (b) ac susceptibility of $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$ at different external pressures. For clarity the $\chi_{\text{ac}}(T)$ data at different pressures have been shifted with respect to each other.

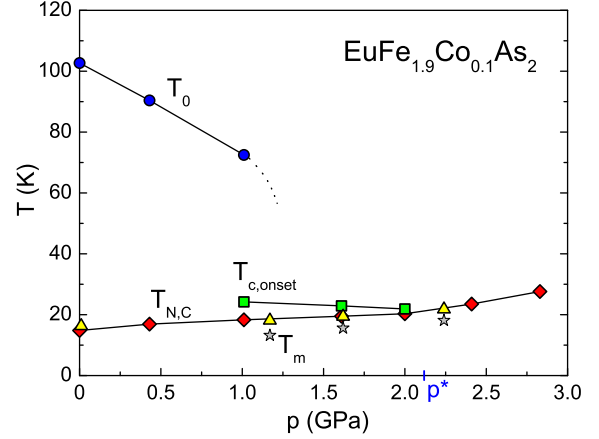


Figure 4. Temperature-pressure phase diagram of $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$.

observed previously, but at higher pressures in EuFe_2As_2 [1]. The drop in $\rho(T)$ as well as the following maximum are much more pronounced at $p = 1.61$ GPa, but are hardly visible anymore at $p = 2$ GPa. At even higher pressures no indication of superconductivity is present in the resistivity data anymore.

The results of the $\chi_{\text{ac}}(T)$ experiments are presented in Fig. 3b. The $\chi_{\text{ac}}(T)$ curve at the lowest pressure ($p = 0.01$ GPa) and the curves at higher pressures are qualitatively different. While the shape of $\chi_{\text{ac}}(T)$ at $p = 0.01$ GPa is reminiscent of an AFM phase transition, the shape at $p > 0.01$ GPa is typical for a FM transition. A change of the magnetic groundstate from AFM to FM has been found in $\text{EuFe}_2\text{Co}_2\text{As}_2$ on P-doping on the As-site too [10]. P doping corresponds to the application of positive chemical pressure, therefore, our finding is not surprising and in agreement with the doping studies. On increasing pressure the anomaly in $\chi_{\text{ac}}(T)$ at $T_{\text{N,C}}$ shifts to higher temperatures [9]. At pressures $p > 0.01$ GPa a second feature appears in $\chi_{\text{ac}}(T)$ below T_{C} at T_{m} [9]. We attribute this kink to a change in the magnetic structure from FM to AFM. No feature in $\rho(T)$ is visible at T_{m} . A detailed study of the magnetic properties will be presented elsewhere. It is important to note that we do not observe a clear signature of superconductivity in our χ_{ac} results.

In our study we observe a less robust superconductivity than reported in literature [6, 7]. This might be related to different sample preparation procedures or to internal strain in the samples. An important influence of internal/external residual strain on the observed physical properties appears also in our experiments. After releasing the pressure, we still find reentrant SC behavior in $\rho(T)$ similar to the data at 1.61 GPa (not shown).

The $T-p$ phase diagram in Fig. 4 summarizes our results. While $T_{\text{C}}(p)$ decreases only slightly with increasing pressure $T_{\text{N,C}}(p)$ increases strongly by a factor of nearly 2 from 14.8 K at ambient pressure to 27.6 K at 2.83 GPa, the highest pressure of our experiment. We find a good agreement

between $T_{N,C}(p)$ obtained from the $\rho(T)$ and $\chi_{ac}(T)$ experiments. The strong increase of $T_{N,C}(p)$ with pressure has not been reported in EuFe_2As_2 [1]. There, the magnetic ordering temperature is nearly independent of pressure. The different pressure dependencies of $T_{c,\text{onset}}(p)$ and $T_{N,C}(p)$ in $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$ lead to a crossing of both temperature lines [$T_{c,\text{onset}}(p) = T_C(p)$] in the $T-p$ phase diagram at a pressure p^* slightly higher than 2 GPa. Above p^* once $T_C > T_c$ no indication of any SC transition is visible in the resistivity data. Already at 2 GPa where $T_c = 21.9$ K and $T_C = 20.3$ K get rather close. The feature associated to the SC transition is considerably reduced compared to the previous pressure.

4. Discussion and Summary

In summary our results show that in the substitution series $\text{EuFe}_{2-x}\text{Co}_x\text{As}_2$ the SDW transition temperature T_0 is suppressed rapidly upon increasing Co concentration, while the ordering temperature of the Eu^{2+} moments is almost constant. For $x = 0.2$ we observe the onset of superconductivity in $\rho(T)$ at ≈ 10 K. For $x < 0.2$ and $x > 0.2$ no indication of superconductivity is present. The strong suppression of T_0 as function of the Co concentration and the appearance of superconductivity in the $T-x$ phase diagram is consistent with the expectation from the comparison with the homolog non- f Co-substitution series $\text{SrFe}_{2-x}\text{Co}_x\text{As}_2$ [5]. However, there superconductivity is stable in a broader concentration range. The presence of only a narrow SC regime in the Eu system might be related to the presence of the local moment Eu^{2+} magnetism.

Pressure rapidly suppresses the SDW instability at T_0 in $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$. In the same pressure region where the signature of T_0 is lost in $\rho(T)$ the onset of superconductivity is observed. At low temperatures our results suggest a strong detrimental effect of the Eu^{2+} magnetism on the superconductivity. In an intermediate pressure range ($1.01 \text{ GPa} \leq T \leq 2 \text{ GPa}$) a the system is on the verge to a SC state, indicated by the drop in the resistivity below $T_{c,\text{onset}}$. However, the formation of the SC state is inhibited by the formation of long range magnetic order of the Eu^{2+} moments. A similar result has been reported previously in undoped EuFe_2As_2 [1]. The important difference in the present study on $\text{EuFe}_{1.9}\text{Co}_{0.1}\text{As}_2$ is that upon increasing pressure $T_{N,C}(p)$ increases while $T_{c,\text{onset}}(p)$ decreases and, therefore, $T_C(p)$ becomes greater than $T_{c,\text{onset}}(p)$ above a certain pressure p^* . Once $T_C(p) > T_{c,\text{onset}}(p)$ no signature of superconductivity is found anymore. We take this as an evidence for the strong detrimental effect of the Eu^{2+} magnetism on the the formation of the SC state. As our ac-susceptibility results indicate Eu^{2+} orders ferromagnetically under pressure. Thus, we speculate that the internal magnetic fields due to the ferromagnetically ordered Eu^{2+} moments suppress the superconductivity in the iron-arsenide layers.

Acknowledgements

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